

Selection of Agricultural Straws as Sustained-Release Carbon Source for Denitrification in a Drawer-Type Biological Filter

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Abstract The aim of this paper is to select feasible agricultural straws as high-quality sustained-release carbon source and examine the effect of determined agricultural organic waste on improving denitrification efficiency. Five kinds of agricultural straws, i.e., the rice straw, the corn straw, the wheat straw, the broomcorn straw, and the reed straw, were evaluated in a selfdesigned drawer-type biological filter. Results showed that the contents of C, H, and N in the five straws were 34.0~41.0%, 4.9~5.4%, and 1.1~1.5% respectively. The highest TOC release capacity of the rice straw was 12.4 \pm 1.3 mg g⁻¹ and the average TOC release of other waste straws ranged from 6.0 to 9.2 mg g^{-1} . The TN release capacities of all the five straws were at a low level, ranging from 0.2 to 1.4 mg g^{-1} . Preliminary denitrification studies showed that the corn and the rice straw could be used as high-quality carbon sources, achieving a COD removal rate of 47.3~50.2% and a TN removal rate of 21.8~24.8% for wastewater with low C/N ratio. The rice straw and the corn straw founctioned both as favorable solid carbon sources and biofilm carriers; the carbon source quality of the corn straw lixivium is more beneficial to microbial utilization. The drawer-type biological filter has showed a good efficiency of

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denitrification for nitrogen removal when using agricultural straws as biofilm carriers.

Keywords Agricultural straws \cdot Low C/N ratio wastewater. Drawer-type biological filter. Denitrification

1 Introduction

Because of the efficiency and the good reliability, biological denitrification is widely used in treating municipal sewage (Fernándeznava et al. [2010](#page-9-0)). However, due to large water consumption and combined drainage system in China, the real municipal sewage has a low C/N ratio below 5.0, which restricts biological denitrification (Hongduck and Sangill [2009;](#page-9-0) Wu et al. [2012\)](#page-10-0). Therefore, adding external organics is necessary in many denitrifying practices (Saeed and Sun [2012](#page-10-0)). It has been reported that external carbon sources such as methanol (Sahinkaya and Dursun [2012](#page-10-0)), ethanol (Qambrani et al. [2013](#page-10-0)), sodium acetate (Tang et al. [2018](#page-10-0)), and polymers (Zhang et al. [2016\)](#page-10-0) have been widely applied, whereas they are generally characterized as high-costly, with complicated operating requirements and high-risk to cause secondary pollution, such as overdosing and nitrite accumulation in effluent (Trois et al. [2010](#page-10-0)). Therefore, selection of appropriate carbon source and suitable controlling methods are crucial for biological nitrogen removal systems.

China is an agricultural country that produces a large amount of agricultural wastes (Peng [2009](#page-10-0)). The average

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annual crop residues yield of Chinese major food crops reached 5.2×10^8 tons during the years 2002–2011, 19% of which were burned directly, which is not only a waste of biological resources, but causing serious air pollution (Yang et al. [2015\)](#page-10-0). Agricultural organic wastes are mainly composed of cellulose, hemicellulose, and lignin (Sun et al. [2016\)](#page-10-0), with a structure of large surface area suitable for the growth of microorganisms (Aslan and Türkman [2005](#page-9-0)); they are very convenient to be utilized as sustainable slow-release carbon sources. Some agricultural organic wastes (wheat straw, maize cob, cottons etc.) have been tested and the results demonstrated that they can be used as substrate amendments for enhancing denitrification (Feng et al. [2017](#page-9-0); Gibert et al. [2008;](#page-9-0) Soares and Abeliovich [1998\)](#page-10-0).

Nevertheless, agricultural organic wastes are also consumable; after a long period of operation, some problems will appear (Chu and Wang [2016](#page-9-0)). For example, comparing to the common biological filters, the agricultural wastes are less convenient to be replaced (Wang and Chu [2016](#page-10-0)); plugging problem in solid-phase denitrification (SPD) is another issue to concern (Wang and Chu [2016\)](#page-10-0). Nitrite can be accumulated if the organic substrate does not fulfill the stoichiometric requirements (Hoover et al. [2016](#page-9-0)); the higher the pH values are, the more likely the accumulation of nitrite would occur (Jianping et al. [2003\)](#page-9-0). On the other hand, if the concentration of the added organic substrate is too high, the residual amount can cause secondary pollution, such as high dissolved organic carbon (DOC) and color in ef-fluent for natural organic substances (Healy et al. [2012](#page-9-0); Katarzyna et al. [2015](#page-9-0)).

Therefore, the first objective of this study was to evaluate the feasibility of agricultural organic wastes used as solid carbon sources and microbial growth carriers. The second objective was to investigate the effect of the selected agricultural organic wastes on improving nitrogen removal. We used a self-designed drawer-type biological filter and a preliminary test was carried out with selected agricultural wastes. In this study, five natural agricultural straws were tested in batch experiments for carbon release capacity, inclusive of the leaching test and elemental analysis. The selected agricultural organic wastes were then added into a self-designed drawer-type biological filter to improve denitrification with a short hydraulic retention time and a small footprint. Performance characteristics of the drawer-type biological filter were also investigated.

2 Materials and Methods

2.1 Experimental Materials

Five common and cheap agricultural straws were collected from the Chongming District, Shanghai, China. The corn straw (CS), the rice straw (ReS), the wheat straw (WS), the reed straw (RdS), and the broomcorn straw (BS) were cut into strips of 2~3 cm. The materials were washed with deionized water, placed in an oven under 65 °C, and dryed for more than 24 h. After being cooled naturally, they were loaded into the sample bags for use.

2.2 Leaching Tests of Agricultural Straws

The agricultural wastes were grounded into powder through a 100-mesh sieve and the Heraeus Elementar Vario MACRO elemental analyzer was used to analyze contents of elements of C, H, and N. Five kinds of straws (5 g, dry weight) were placed in an airtight conical flask with 400 mL deionized water and incubated at 25 °C, 150 r min⁻¹ for 15 days. After that, 25 mL of liquid was sampled by a syringe and sent to ICP-MS (7500 inductively coupled plasma mass spectrometer, USA) to analyze the concentration of metals such as Cu, Pb, Cr, Zn, and Ni.

The sterilized agricultural straws were put into an autoclave with the condition pressure of 0.1 Mpa and the temperature of 121 \degree C for 2 h (2.5 g, dry weight), with 100 mL of deionized water put into 250 mL of conical flasks. The lixivium was poured out and replaced by 100 mL of fresh deionized water every 24 h, while liquid samples were taken out for TOC and TN analyses. Two parallel samples were determined for each test.

2.3 The Preliminary Experiment

Three sets of pre-experiments were carried out to test static denitrification of straws with 10 g of CS and RS, and CK (not added carbon source) put in a 500 mL of round bottle respectively. The same amount of sand covering material was put on all the tree sets of experiments preventing fitering materials from floating. Fifty milliliters of the cultured denitrification activated sludge and 400 mL of the nutrient solution with COD of 15.2~25.5 mg L⁻¹, NO₃⁻-N of 25~30 mg L⁻¹, TP of 0.8~1.2 mg L^{-1} , and pH of 6.6~7.5 were added to each

experimental set. The nutrient solution was changed and sampled every day $(24 h)$.

2.4 Experimental Device and Operational Conditions

A self-designed drawer-type biofilter was used as the experimental device (Fig. 1). The system is made of 5 mm thick plexiglass, which size is length (L) 110 mm \times width (W) 110 mm \times height (H) 310 mm. The effective volume is 3 L and the volume of the reaction zone is 1.5 L. The reaction zone is provided with three drawers, which size is L 100 mm \times W $100 \text{ mm} \times H$ 50 mm. Each drawer unit is covered with a rubber ring. Before the device runs, the drawer unit is fixed by screws to seal the main body from air, which ensure the anoxic conditions of the device. The whole device consists of three parts: the water distribution system, the drawer unit, and the slow-release carbon filler. The water distribution system is composed of an inlet valve, a diverter pipe, and four circular ducts. One side of the diverter pipe is provided with four circular ducts which are connected with each other at equal intervals on both sides of the round ducts.

The reactor was fed with the synthetic wastewater, which is simulated as urban sewage plant secondary effluent (Table 1). The specific composition of the test water was shown in Tables 2 and [3.](#page-3-0)

Fig. 1 Diagram of the drawer-type biofilter device, where (1- Vent; 2-Water distribution pipe; 3-Flange; 4-Shunt; 5-Water distribution hole; 6-water inlet valve; 7-Nut; 8-Inlet pipe; 9-Handle; 10- Rubber ring; 11-Slow-release carbon source filler; 12-Drawer device; 13-Sand particles; 14-Support plate; 15-Water outlet; 16- Peristaltic pump)

The inoculated sludge used in the experiment was collected from the anaerobic compartment of a sewage treatment plant in Shanghai with a MLSS concentration of 2.8 \pm 0.2 g L⁻¹; the sludge was inoculated into the filter directly. Flow rate of the filter was 0.02 L min⁻¹ and the empty bed hydraulic retention time was 24 min.

2.5 Analytical Methods

Water quality parameters such as TN, COD, and MLSS were determined with standard methods listed in Table [4;](#page-3-0) TOC was tested with a total organic carbon/total nitrogen analyzer (multi N/C 2100S); while metal content of the leaching liquid was analyzed with an ICP-MS 7500 Inductively Coupled Plasma Mass Spectrometer and the content of C, H, and N elements were determined with a Heraeus Elementar Vario MACRO elemental analyzer.

3 Results and Discussion

3.1 Composition and Elemental Analysis of Five Agricultural Straws

The content of solid elements can reflect the basic background values of carbon and nitrogen contained in agricultural straws. It not only reflects the release capacity of carbon and nitrogen from straws as packing materials, but also their potential risks as secondary pollutants. The contents of C, H, and N in the five kinds of straws indicated that the content of carbon elements was significantly higher than that of hydrogen and nitrogen elements in the study straws (Fig. [2\)](#page-4-0). Among them, the content of the carbon element in the reed straw was the

Table 2 Composition of influent

Composition	Content $(g L^{-1})$
C_6H_1 ₂ O ₆	0.7
KNO ₃	0.15
KH_2PO_4	0.015
NH ₄ Cl	0.01
Trace elements (mL/L)	1

Table 3 Composition of trace elements

Composition	Content (mg L^{-1})
ZnSO ₄ .7H ₂ O	0.12
FeCl ₃	0.9
$Na2MoSO4·2H2O$	0.06
MnCl ₂ ·4H ₂ O	0.06
KI	0.18
CuSO ₄ ·5H ₂ O	0.03
CaCl ₂ ·6H ₂ O	0.15
H_3BO_3	0.15

highest, reaching 41.0%; while the content range of other straw carbon elements was between 34.0 and 38.6%. There was no significant difference in hydrogen and nitrogen content of the five straws. Among them, the mean value of hydrogen is 5.2%, while that of nitrogen is 1.3%.

The contents of Cu, Pb, Cr, Zn, and Ni in each straw extract were tested and the results were shown in Table [5.](#page-4-0) The content of Zn was the highest and Cr was the lowest after 15 days of operation. The content of Pb in two straw leaching solutions was slightly higher than that of the Chinese IV water quality standard value for surface water. Considering the fact that the water flow and velocity are much greater than experimental conditions, such straw will not affect the environmental quality if they are used as biological denitrification carbon source.

Table 4 Analytical methods

Method
Basic potassium persulfate digestion UV spectrophotometry (HJ 636-2012)
Nessler's reagent spectrophotometry (HJ 535-2009)
NO_3 ⁻ -N Ultraviolet spectrophotometry (HJ/T 346-2007)
$NO2--N N- (1-naphthyl) -ethylenediamine photometry$ $(GB7493-87)$
Ammonium molybdate spectrophotometric method(GB11893-89)
Rapid digestion spectrophotometry (HJ/T 399-2007)
pH meter HE20 (METTLER TOLEDO Instrument Co., Ltd.)
Haschi dissolved oxygen analyzer
Gravimetric method

3.2 Carbon and Nitrogen Release Capacity

As shown in Fig. [3](#page-5-0)a, TOC concentrations of five kinds of straw lixivium reached the maximum level on the first day, among which the rice straw reached the highest release value at 4.0 ± 1.3 mg g⁻¹. The release of the corn straw, the wheat straw, and the broomcorn straw was in the range of 6.0~9.2 mg g^{-1} ; while the lowest release of carbon was from the reed straw at 1.8 ± 0.4 mg g⁻¹. Five different straws released a certain amount of carboncontaining dissolved organic matter within the first 3 days and then reached a stable low level. The process of releasing dissolved organic matter by solid carbon source materials can be divided into two stages, i.e., fast release in the first stage and slow release in the late stage. At the beginning stage, the initial concentration of COD in water increased rapidly and reached the maximum value due to the release of easily decomposable carbohydrates and rapid degradation of watersoluble substances (Zhou et al. [2008\)](#page-10-0); carboncontaining small molecules attached to the surface of the material and the easily soluble small molecules detached out of the material gradually, as it expanded quickly and was dissolved into the aqueous solution, thus exhibiting a rapid rise of COD concentration in the water (Shao et al. [2011](#page-10-0)). Then, as the carbon-containing small molecules inside the carbon source materials were further decomposed, dissolved, and released into the water body, hardly decomposable substances such as hemilignin and lignin accumulated, while the simple decomposition of the material was inhibited. Therefore, the decomposition rate slowed down gradually and achieved a balance state (Shao et al. [2011](#page-10-0); Yang et al. [2015](#page-10-0)). The aim of the experiment is to provide sufficient carbon source for low C/N ratio wastewater denitrification; CS and RS are the target materials based on the preliminary experiments.

As shown in Fig. [3b](#page-5-0), TN release from the five kinds of straw lixivium was at a low level, ranging from 0.2 to 1.4 mg g−¹ , with the highest release values for CS and ReS at 1.3 ± 0.2 mg g⁻¹ and 1.3 ± 0.3 mg g⁻¹ respectively. TN release for most straws reached a certain platform in the first 3 days. The highest TN release occurs on the first day and then gradually decreased with time, because the straw is composed of cellulose, hemi-lignin, and lignin, which has active groups to adsorb nitrogenous substances (Orlando et al. [2002\)](#page-10-0). During the stabilization stage, some stalks increased greatly in terms of nitrogen release, which was caused

Fig. 2 Elemental analysis of five straws

by the release of nitrogenous protein during soaking and the hydrolysis of protein resulted in an increase of TN concentration (Li and Hao [2013a\)](#page-10-0).

Figure [3](#page-5-0)c showed the overall trend of C/N ratio of the five different straws lixivium. The hydrolysis of small molecule substances attached to the surface of solid carbon source could bring about a gradual increase of the C/N ratio (Li and Hao [2013a\)](#page-10-0); the hydrolysis of protein substances, however, could release nitrogen and reduce the C/N ratio gradually. Among the five kinds of straws lixivium, the C/N ratio was between 5.0 and 13.0. Zhong et al. found that when the hydrolysate of plant material was used as carbon source, the C/N ratio of the hydrolysate was much higher than the appropriate C/N ratio of denitrification for wastewater (Zhong et al. [2014\)](#page-10-0). The C/N ratio in the bulk water can reach 9.0 after adding carbon source and achieve high efficiency of denitrification. In this study, the average C/N ratio of CS and ReS lixivium were above 10.0. Therefore, two different straws can further increase C/N

ratio, provide continuous supply of carbon source, and have good carbon release stability.

3.3 Static Denitrification Pre-experiment

Results of the static denitrification pre-experiments with CS added, RS added, and CK (without carbon source added) were demonstrated in Fig. [4.](#page-6-0) Concentration of NO₃⁻-N decreased firstly and then remained identical to the influent in CK. Due to insufficient carbon source in the influent, the denitrifying bacteria consumed their own carbon source for metabolism in the first 2 days, and then large numbers of denitrifying bacteria died and effluent NO_3 ⁻-N is maintained identical to the influent. In other groups with carbon source dosage, the effluent removal rate of NO_3^- -N reached 50% and the effluent was about $10.4-17.2$ mg·L⁻¹. Carbon source dosage could effectively promote denitrification for low C/N wastewater.

Fig. 3 a TOC concentration, b TN concentration, and c C/N value in lixivium of the five different straws

3.4 Characterization of COD Removal in Drawer-Type Biofilter with CS and ReS

Figure [5](#page-7-0) indicated that effluent COD decreased rapidly from the first day to the second day. A large amount of molecular substances lead to high effluent COD concentration in the first days and degrade under the action of denitrifying bacteria, resulting in decrease of effluent COD (Cameron and Schipper [2012](#page-9-0)). Robertson also found the high release of dissolved organic carbon (DOC) in the effluent, especially during the start-up period (Robertson [2010\)](#page-10-0). Effluent COD using two kinds of straws decreased rapidly after a short initial increasing period, indicating the biofilm attached on granules gradually got mature with a short time. Effluent COD using CS and ReS were 24.4 ± 4.8 mg L⁻¹ and $30.6 \pm$ 3.5 mg L^{-1} respectively. The COD removal rate was about 51.4% with CS as filter and 38.7% with ReS

relatively. During the start-up period, the effluent COD concentration is even higher than the influent due to excessive carbon release from the straws. In order to avoid excessive COD production at the beginning of the experiment, the filters can be soaked 2 or 3 days before used (Li and Hao [2013b\)](#page-10-0). Values of fluorescence peak of excitation $(E_x)/$ emission (E_m) wavelength are indicators of certain organic matters (Hudson et al. [2010](#page-9-0)). Figure [6](#page-7-0) indicates that the released organics from the CS filter are mainly tryptophan with $E_x/E_m = 250 \approx 275/325 \approx 350$ and fulvic acid with $E_x/E_m = 325 \sim 350/400 \sim 500$; similarly, fluorescence peaks characterzing tryptophan and fulvic acid are also observed in the ReS filter. Fulvic acid is a kind of humic acid and has strong biological inertia. Tryptophan is a readily degradable substance and is one of the important amino acids that make up proteins; the carbon source quality of CS leaching solution is more beneficial to microbial utilization (Hudson et al. [2010\)](#page-9-0).

Fig. 4 Comparison of NO_3^- -N concentration in the static denitrification pre-experiment

3.5 Denitrification Efficiency in the Drawer-Type Biofilter with CS and ReS

As demonstrated in Fig. [7](#page-8-0)a, the curve of effluent $NO₃⁻$ N showed a decreasing trend. At the beginning of the study, the removal effect of $NO₃⁻-N$ was not obvious. Wong et al. (Wong and Lee [2011](#page-10-0)) concluded that the theoretical value of C/N (COD/ NO_3 ⁻-N) required for complete denitrification was 2.86. When the C/N ratio was greater than 5.0, the denitrification effect was obvious in practical applications and the optimal range of C/ N ratio was between 5.0 and 10.0 (Zhao et al. [2011\)](#page-10-0). The effluent NO_3 ⁻-N using CS and ReS was $11.0 \pm$ 0.5 mg L⁻¹ and 11.5 ± 0.6 mg L⁻¹ respectively, while the removal rates were $22.3 \pm 3.1\%$ and $20.1 \pm 2.5\%$ respectively. Drawer-type filters could perform slow denitrification; the reasons for the low denitrification efficiency may be as follows: on the one hand, the wastewater is not in full contact with the microorganisms and the hydraulic retention time is relatively short (about 30 mins) (Hoover et al. [2016;](#page-9-0) Wang and Chu [2016\)](#page-10-0); on the other hand, the influent total nitrogen concentration is relatively low and further removal of total nitrogen is difficult; it may requires better environmental conditions and external forces, such as electrical stimulation. Researchers have studied the interspecific electron transport of electroactive microorganisms and denitrifying bacteria.

It is found that G. sulfurreducens, which cannot utilize nitrates, can accelerate the formation of denitrifying microbial communities, eliminate the lag phase, and slow the accumulation of nitrous oxide, speed up the denitrification rate (Wan et al. [2018\)](#page-10-0). NH3-N concentration did not change much and the removal effect was not obvious (Fig. [7b](#page-8-0)), because the device performed denitrification under the interaction of anoxic condition and denitrifying bacteria. Nitrite concentration was very low in Fig. [7](#page-8-0)d (less than 0.05 mg·L⁻¹); no accumulation of nitrite was observed. Figure [7c](#page-8-0) showed that there were linear relationship between effluent TN and NO_3 ⁻-N (p < 0.05). The effluent TN using CS and ReS was 12.1 ± 0.6 mg L⁻¹ and 12.6 ± 0.8 mg L⁻¹ respectively, while removal rates were about $24.3 \pm 1.2\%$ and $22.1 \pm 1.5\%$ respectively.

Carbon source is the main factor limiting denitrification (Li et al. [2012\)](#page-10-0). In the initial stage of the study, the carbon-containing small molecular substances attached to surface of the material and easily soluble small molecular substances inside the material were rapidly dissolved into the aqueous solution, resulting in high effluent COD (Zhou et al. [2008\)](#page-10-0). Then, hydrolysis of carboncontaining small molecular substances in the filter was hindered. Moreover, the contact time of the aqueous solution with the biofilm was too short, resulting in insufficient denitrification. The type of carbon sources was also one of the main factors affecting the denitrification (Lee et al. [2009\)](#page-9-0); for example, the small molecule carbon source could be directly used by denitrifying bacteria (Yin et al. [2009](#page-10-0)). Since the contact time between the aqueous solution and the biofilm was short,

Fig. 5 Effluent COD concentration from the drawertype bio-filter with two different straws

the microorganism could only use the small molecular carbon source in the aqueous solution, while the macromolecular carbon source released by the filler was not used, resulting in poor denitrification. However, when wastewater contains small molecular carbon source, denitrifying bacteria can rapidly multiply and promote the release of the slow-release carbon source, producing a co-metabolic effect that not only enables denitrifying bacteria multiplying rapidly and denitrify efficiently, but also promotes sustained release of carbon sources contents in the straw filters. Adding some amount of small molecule carbon source can promote the large-scale reproduction of denitrifying bacteria with the slow release of carbon source from the straws, which is valuable to the scale up and application of the process.

As can be seen from Fig. [8,](#page-8-0) approximately only 10% of TP is removed using CS and ReS; there is no significant difference between CS and ReS $(p > 0.05)$. The TP removal rate shows a trend of decreasing after rising, which was different from the observation of Tang et al. who concluded that, during the start-up period of experiment, excessive organic matter concentration enhanced the competition between heterotrophic bacteria and denitrifying phosphate accumulating organisms (DPAOs); the reaction rate of DPAOs is lower than that of heterotrophic bacteria, and the phosphorus absorption performance of DPAOs is suppressed, making TP

Fig. 6 3D-EEM of water-leach liquor from two different straws

Fig. 7 a NO₃⁻-N, b NH₃-N, c TN, and d NO₂⁻-N concentration of effluent from drawer-type bio-filter with two different straws

removal rate reduced (Tang et al. [2016\)](#page-10-0). However, the organic matter concentration in this experiment is not very high, while the growth and metabolism of DPAOs are not affected by other heterotrophic bacteria. As a result, TP removal rates increases monotonically with time. When the influent C/N is higher than the optimum C/N suitable for DPAOs, the activity of DPAOs is suppressed (Tang et al. [2016\)](#page-10-0).

Fig. 8 TP removal efficiencies of the drawer-type bio-filter with two different straws: a CS, b ReS

When the process is scaled up, the issues need to be more comprehensive to ensure that the reactor can operate normally and sustainably, which has guiding significance for practical engineering applications. The following measurements and/or concerns should be considered carefully when the process is scaled up. (1) The bottom of the drawer unit needs a hard material layer that is resistant to acid, alkali, corrosion, and chemical reaction, which prevents the filter from falling due to gravity during operation. (2) Online detectors (dissolved oxygen, pH value, reduction potential, conductivity, temperature, etc) should be installed at the inlet and outlet of the reactor; when there is a problem with the relevant indicators, it can automatically alarm. (3) A backwashing system should be installed inside the reactor. (4) The filter needs to be soaked for 2–3 days before use, preventing the deterioration of water quality from excessive release of the carbon source. (5) The filter needs to be replaced in time after failure, and the replacement cycle is about 30–45 days; the replaced filter can be dried, crushed, and applied to farmland as an organic fertilizer; (6) According to the theory and the actual operation of the laboratory scale process, the sewage treatment plant of 1 million inhabitants needs to dose around 100 tons of slowly released straws on the daily average.

4 Conclusions

Compared with other agricultural straws, the corn straw and the rice straw were found to have more carbon and less nitrogen release capacity; thus, the ratio of C/N is higher. The carbon source quality of the CS leaching solution is better; it is the more favorable solid carbon source and biofilm carrier. During the long period of operation, the whole drawer-type biological filter is not plugged and no nitrite accumulation is observed. Therefore, the drawer-type biological filter has a good efficiency in denitrification for nitrogen removal when using agricultural straws as biofilm carriers.

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Compliance with Ethical Standards

Conflict of Interest The authors declare that they have no conflict of interest.

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References

- Aslan, Ş., & Türkman, A. (2005). Combined biological removal of nitrate and pesticides using wheat straw as substrates. Process Biochemistry, 40, 935–943.
- Cameron, S. G., & Schipper, L. A. (2012). Hydraulic properties, hydraulic efficiency and nitrate removal of organic carbon media for use in denitrification beds. Ecological Engineering, 41, 1–7.
- Chu, L., & Wang, J. (2016). Denitrification of groundwater using PHBV blends in packed bed reactors and the microbial diversity. Chemosphere, 155, 463.
- Feng, L., Chen, K., Han, D., Zhao, J., Lu, Y., Yang, G., Mu, J., & Zhao, X. (2017). Comparison of nitrogen removal and microbial properties in solid-phase denitrification systems for water purification with various pretreated lignocellulosic carriers. Bioresour Technology, 224, 236–245.
- Fernándeznava, Y., Marañón, E., Soons, J., & Castrillón, L. (2010). Denitrification of high nitrate concentration wastewater using alternative carbon sources. Journal of Hazardous Materials, 173, 682.
- Gibert, O., Pomierny, S., Rowe, I., & Kalin, R. M. (2008). Selection of organic substrates as potential reactive materials for use in a denitrification permeable reactive barrier (PRB). Bioresource Technology, 99, 7587–7596.
- Healy, M. G., Ibrahim, T. G., Lanigan, G. J., Serrenho, A. J., & Fenton, O. (2012). Nitrate removal rate, efficiency and pollution swapping potential of different organic carbon media in laboratory denitrification bioreactors. Ecological Engineering, 40, 198–209.
- Hongduck, R., & Sangill, L. (2009). Comparison of 4-stage biological aerated filter (BAF) with MLE process in nitrogen removal from low carbon-to-nitrogen wastewater. Environmental Engineering Science, 26, 163–170.
- Hoover, N. L., Bhandari, A., Soupir, M. L., & Moorman, T. B. (2016). Woodchip denitrification bioreactors: Impact of temperature and hydraulic retention time on nitrate removal. Journal of Environmental Quality, 45, 803.
- Hudson, N., Baker, A., & Reynolds, D. (2010). Fluorescence analysis of dissolved organic matter in natural, waste and polluted waters—a review. River Research & Applications, 23, 631–649.
- Jianping, W., Lei, P., Liping, D., & Guozhu, M. (2003). The denitrification treatment of low C/N ratio nitrate-nitrogen wastewater in a gas–liquid–solid fluidized bed bioreactor. Chemical Engineering Journal, 94, 155–159.
- Katarzyna, B., Dorota, K., & Żuchniewski, K. (2015). Glycerine as a carbon source in nitrite removal and sludge production. Chemical Engineering Journal, 267, 324–331.
- Lee, C. G., Fletcher, T. D., & Sun, G. (2009). Nitrogen removal in constructed wetland systems. Engineering in Life Sciences, 9, 11–22.
- Li, B., & Hao, R.-X. (2013a). Comparison and optimization of cellulose carbon source for denitrification filter. Environmental Science, 34, 1428–1434.
- Li, B., & Hao, R. X. (2013b). Comparison and optimization of cellulose carbon source for denitrification filter. Environmental Science, 34, 1428–1434.
- Li, G., Chen, J., Yang, T., Sun, J., & Yu, S. (2012). Denitrification with corncob as carbon source and biofilm carriers. Water Science & Technology, 65, 1238–1243.
- Orlando, U. S., Baes, A. U., Nishijima, W., & Okada, M. (2002). A new procedure to produce lignocellulosic anion exchangers from agricultural waste materials. Bioresour Technology, 83, 195–198.
- Peng, J. (2009). Review and discussion on utilization of agricultural waste resources in China. Ecology and Environmental Sciences, 18, 794–798.
- Qambrani, N. A., Jung, S. H., Yong, S. O., Yong, S. K., & Oh, S. E. (2013). Nitrate-contaminated groundwater remediation by combined autotrophic and heterotrophic denitrification for sulfate and pH control: batch tests. Environmental Science & Pollution Research, 20, 9084–9091.
- Robertson, W. D. (2010). Nitrate removal rates in woodchip media of varying age. Ecological Engineering, 36, 1581–1587.
- Saeed, T., & Sun, G. (2012). A review on nitrogen and organics removal mechanisms in subsurface flow constructed wetlands: dependency on environmental parameters, operating conditions and supporting media. Journal of Environmental Management, 112, 429–448.
- Sahinkaya, E., & Dursun, N. (2012). Sulfur-oxidizing autotrophic and mixotrophic denitrification processes for drinking water treatment: elimination of excess sulfate production and alkalinity requirement. Chemosphere, 89, 144–149.
- Shao, L., Wang, S., Jin, W., & Yin, H. (2011). Performance of new solid carbon source materials for denitrification. Environmental Science, 32, 2323–2327.
- Soares, M. I. M., & Abeliovich, A. (1998). Wheat straw as substrate for water denitrification. Water Research, 32, 3790–3794.
- Sun, S., Sun, S., Cao, X., & Sun, R. (2016). The role of pretreatment in improving the enzymatic hydrolysis of lignocellulosic materials. Bioresour Technology, 199, 49–58.
- Tang, J., Huang, P., Jinxiang, F. U., Liu, J., Chen, J., You, K., Yuan, Y. (2016). Effect of corncob as external carbon source on nitrogen and phosphorus removal of SBBR. Chinese Journal of Environmental Engineering, 10, 4705–4710.
- Tang, J., Wang, X. C., Hu, Y., Pu, Y., Huang, J., Hao, N. H., Zeng, Y., & Li, Y. (2018). Nitrogen removal enhancement using lactic acid fermentation products from food waste as external carbon sources: performance and microbial communities. Bioresource Technology, 56, 259–268.
- Trois, C., Pisano, G., & Oxarango, L. (2010). Alternative solutions for the bio-denitrification of landfill leachates using pine bark and compost. Journal of Hazardous Materials, 178, 1100– 1105.
- Wan, Y., Zhou, L., Wang, S., Liao, C., Li, N., Liu, W., & Wang, X. (2018). Syntrophic growth ofGeobacter sulfurreducens accelerates anaerobic denitrification. Frontiers in Microbiology, 9, 1572.
- Wang, J., & Chu, L. (2016). Biological nitrate removal from water and wastewater by solid-phase denitrification process. Biotechnology Advances, 34, 1103–1112.
- Wong, B. T., & Lee, D. J. (2011). Denitrifying sulfide removal and carbon methanogenesis in a mesophilic, methanogenic culture. Bioresource Technology, 102, 6673–6679.
- Wu, W., Yang, F., & Yang, L. (2012). Biological denitrification with a novel biodegradable polymer as carbon source and biofilm carrier. Bioresource Technology, 118, 136–140.
- Yang, X. L., Jiang, Q., Song, H. L., Gu, T. T., & Xia, M. Q. (2015). Selection and application of agricultural wastes as solid carbon sources and biofilm carriers in MBR. Journal of Hazardous Materials, 283, 186–192.
- Yin, F. F., Wang, S. Y., Ang, X. Y., Hou, H. X., Peng, Y. Z., & Wang, W. (2009). Effects of carbon source types on denitrification performance at low temperature. Environmental Science, 30, 108.
- Zhang, Q., Ji, F., & Xu, X. (2016). Effects of physicochemical properties of poly-ε-caprolactone on nitrate removal efficiency during solid-phase denitrification. Chemical Engineering Journal, 283, 604–613.
- Zhao, Y. J., Hui, Z., Chao, X., Nie, E., Li, H. J., He, J., & Zheng, Z. (2011). Efficiency of two-stage combinations of subsurface vertical down-flow and up-flow constructed wetland systems for treating variation in influent C/N ratios of domestic wastewater. Ecological Engineering, 37, 1546–1554.
- Zhong, S., Yang, Y., Ran, T., Li, L., Min, Z., & Zhao, J. (2014). Carbon releasing characteristics and denitrification effects of five plant materials. Chinese Journal of Environmental Engineering, 8, 1817–1824.
- Zhou, G. Z., Sun, J., Zhang, X., & Guang-He, L. I. (2008). Carbon source study on biological denitrification in groundwater. Environmental Science & Technology, 31, 4–6.